LLNL-PROC-450382

LAWRENCE LIVERMORE NATIONAL LABORATORY

LLNL RADIATION DETECTOR MATERIALS FOR INTERNATIONAL SAFEGUARDS

S. A. Payne, N. J. Cherepy, R. Nikolic, N. Zaitseva, A. Conway, T. F. Wang, A. Dougan

August 20, 2010

Institute of Nuclear Materials Management Conference
Baltimore, VA, United States
July 11, 2010 through July 15, 2010

LLNL RADIATION DETECTOR MATERIALS FOR INTERNATIONAL SAFEGUARDS

Stephen Payne, Nerine Cherepy, Rebecca Nikolić, Natalia Zaitseva,
Adam Conway, Tzu-Fang Wang, and Arden Dougan
Lawrence Livermore National Laboratory
P.O. Box 808, Livermore, CA 94551

ABSTRACT
LLNL's campaign for developing advanced radiation detector materials has been accelerated over the last few years. We have pursued many different technologies including: scintillator crystals, ceramics, and polymers; thermal neutron detectors based on a boron-loaded silicon platform; organic high-energy neutron detectors; and the semiconductors TlBr and CZT with specialized contacts. In this paper, we discuss these technologies and future Safeguards needs.

SAFEGUARDS APPLICATIONS
The goal of the Next Generation Safeguards Initiative is to strengthen international safeguards through the development and application of tools, technologies, and methods to optimize the effectiveness and efficiency. Applications identified include detectors for sensing neutron multiplicity of fissile material, enrichment assessments, precise isotopic measurements, alternatives to helium-3, and deployments in harsh environments, see Fig. 1. In the next section we describe the new detector materials that are applicable to these uses.

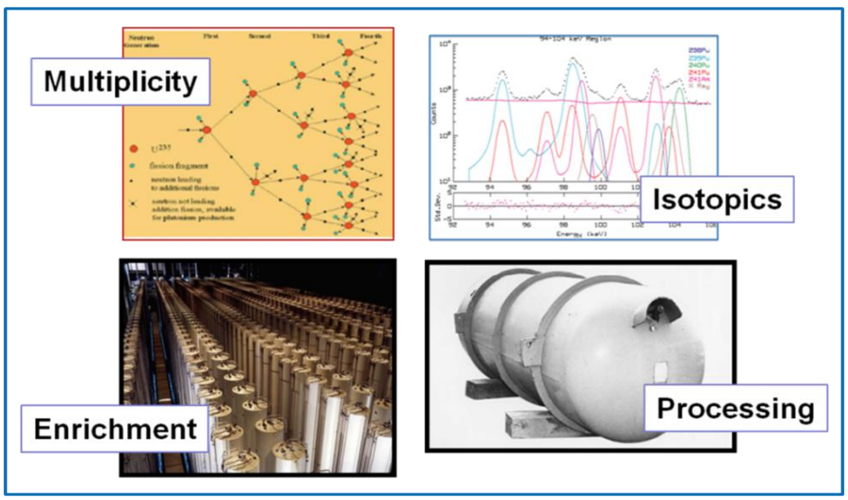

Fig. 1: Depiction of processes and measurements of crucial importance to safeguards, including neutron multiplicity, isotopic determinations, enrichment, and chemical processing.

NEW LLNL DETECTOR MATERIALS
Having ready access to high-energy neutron detectors based on organic crystals would be extremely valuable, since single crystals offer better neutron/gamma discrimination compared to liquid scintillators, do not present a fire hazard, and provide better time resolution than thermal neutron detectors. The present best-known crystalline candidate is melt-grown stilbene, which has limited commercial availability at this time. Therefore, we have been pursuing an alternative approach based on use of solution-growth; examples of crystallizers and single crystals are shown in Fig. 2.[1] We have grown large crystals of stilbene and crystals mixed with stilbene (e.g. diphenyl acetylene)

that offer equivalent or superior performance compared to commercial melt-grown stilbene. Solution grown organic crystals appear to be a viable alternative to melt growth and a pathway for greatly improving the commercial availability of this class of materials, since solution growth is simpler, less costly, and does not introduce stresses into the material.

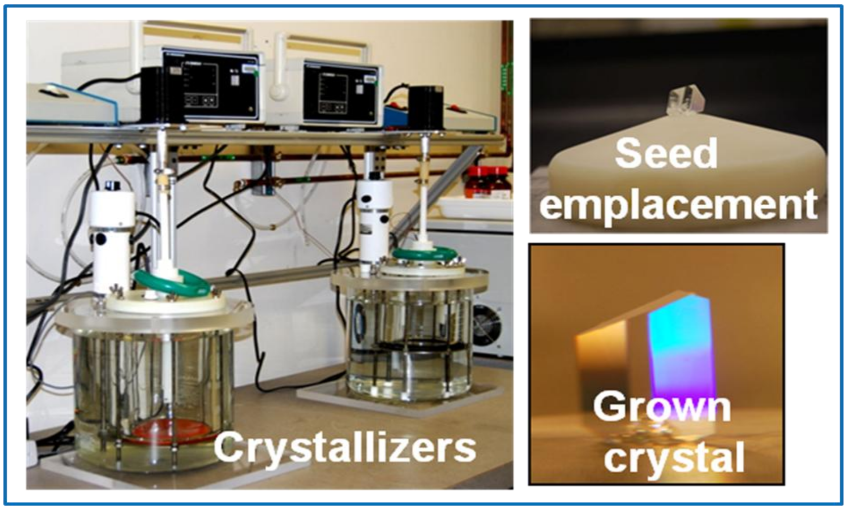

Fig. 2: Equipment and crystals based on the solution-growth technique. Crystals of 2-inch size have been grown, opening up a new avenue for producing organic crystals such as stilbene by an inexpensive growth route.

We have also been developing a solid-state pathway to the detection of thermal neutrons, to serve as a replacement for the traditional ^{3}He detectors. We have developed a boron-loaded thermal neutron detector based on a silicon platform (Fig. 3),[2] wherein the silicon is fabricated into so-called "pillar" structure that is two microns wide at a pitch of two microns. This technology has been developed with an eye toward utilizing the existing commercial semiconductor infrastructure to enable widespread manufacturing of this device in the future. Elemental boron fills the regions between the silicon pillars (having been deposited by chemical vapor deposition, CVD). The boron depth is 20 to 50 microns to assure efficient interaction with the thermal neutrons, while the transverse widths are 2 microns to enable the nuclear reaction products to emerge from the boron at high energy and thereby impinge on the silicon pillars where electrons/holes are generated and registered in the external circuitry.

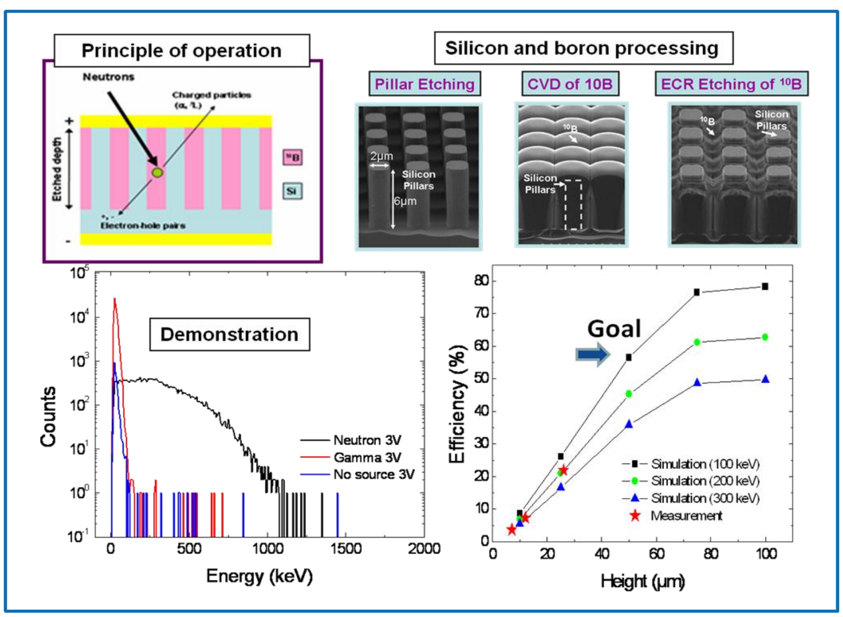

Fig. 3: The "pillar" thermal neutron detector, based on a three-dimensional silicon structure, where boron is deposited between the silicon pillars by chemical vapor deposition, which is then etched back to apply contacts. Excellent (10^5) neutron/gamma discrimination is obtained. The demonstration of 22% efficiency is plotted; the intent is to achieve >50% efficiency using 50 micron pillars in the future.

We have developed two new types of scintillator detectors, based on the single crystal SrI_2(Eu) and the transparent ceramic garnet GYGAG(Ce).[3] From the data on these materials in Fig. 4 and the comparisons with the commercially available materials $LaBr_3$(Ce) and NaI(Tl), we can see that SrI_2(Eu) offers breakthrough resolution, while the gadolinium-yttrium-gallium-aluminum garnet (GYGAG) doped with cerium has fairly good resolution and additionally is mechanically robust such that it can be deployed in harsh environments. Shown in Fig. 4 is melt-grown SrI_2(Eu) crystal in a hermetically sealed enclosure, and a GYGAG(Ce) ceramic produced from nanoparticles that are first formed into a green body, then calcined, vacuum-sintered, and hot isostatically pressed to transparency. Lastly, we mention in passing that we are developing polymers loaded with organometallic bismuth compounds and have been successful in inducing a strong photopeak in this class of materials; the current resolution is 15% at 662 keV which is likely to be improved in the future.

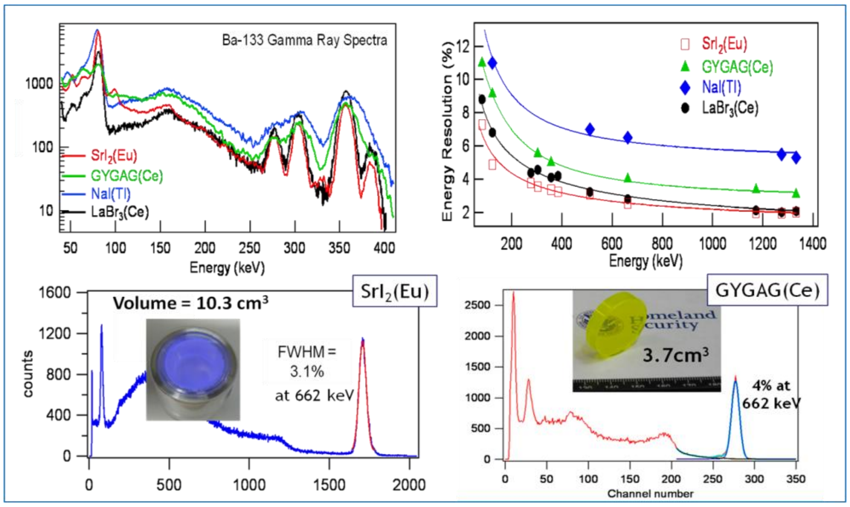

Fig. 4: We have been developing two new scintillator materials: single crystal SrI_2(Eu) which affords high resolution, and the transparent ceramic based on a mixture of Gd-Y-Ga-Al oxides, to provide good resolution as well as phase stability to yield large size with mechanical robustness.

There is high leverage for improved CZT detectors in the development of superior electrical contacts with both electron- and hole-blockers,[4] since this would reduce the leakage current as shown in Fig. 5. Our pathway has been to include an amorphous silicon layer in the structure with gold or platinum contacts. Using controlled conditions (albeit with relatively low resistivity CZT in this study), we were able to show that improvement in electrical performance could be achieved, although further work is required to fully develop this approach with high-quality CZT and with optimized surface polishing, etching and passivation.

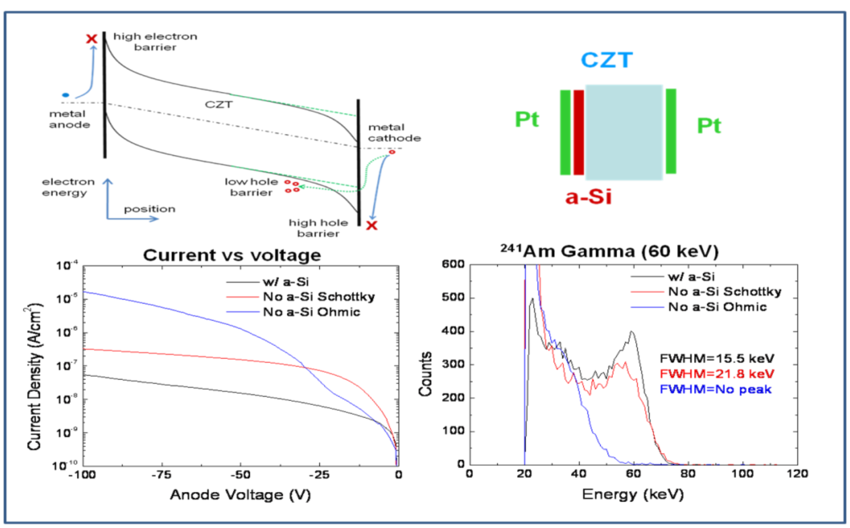

Fig. 5: Development of CZT contacts with improved leakage current on the basis of incorporating amorphous silicon within the contact structure, revealing that improved resolution can be achieved with blocking contacts.

We are also working with RMD, Inc.[5] in the development of alternative semiconductor options, particularly TlBr. Here we are developing specialized contacts to enable a so-called field-annealing procedure in order to reduce the ionic motion at room temperature so that long-term stability is achieved. RMD has achieved resolution approaching 1% at 662 keV.

SUMMARY

We described our work in new materials and fabrication techniques, as it relates to safeguards needs. In particular, we discussed the use of solution-grown organic crystals for high-energy neutron detection for multiplicity registration, the development of a boron-loaded "pillar" silicon platform for thermal neutron detection as an alternative to ^{3}He tubes, a high-resolution $SrI_2(Eu)$ scintillator for enrichment assessments, the use of a robust GYGAG(Ce) transparent ceramic for use in harsh environments, and a pathway for potentially improving the resolution of CZT based on superior contacts.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. We wish to thank NA22 of the Department of Energy and DNDO of the Department of Homeland Security for their financial support and interest in this work. We wish to thank the many staff at LLNL and at other institutions for their help in executing these projects, including: Kanai Shah and coworkers at RMD; Arnold Burger and coworkers at Fisk University; Lynn Boatner and coworkers at ORNL; Lorenzo Fabris and coworkers at ORNL; Barry Cheung and coworkers at the University of Nebraska; and, at LLNL, P Beck, L Carmen, O Drury, M Faust, S Fisher, A Glenn, R Graff, S Hamel, J Kuntz, A Nelson, C Reinhardt, J Roberts, B Rupert, R Sanner, Z Seeley, Q Shao, S Sheets, B Sturm, P Thelin, and L Voss.

REFERENCES

1. N Zaitseva, J Newby, S Hamel, L Carman, M Faust, V Lordi, N Cherepy, W Stoeffl, and S Payne, "Neutron detection with single crystal organic scintillators," Proc. SPIE, 7449, 7449-11 (2009).
2. R Nikolić, A Conway, C Reinhardt, R Graff, T-FWang, N Deo, and C Cheung, "Pillar structured thermal neutron detector," 2008 9TH International Conference on Solid-State and Integrated-Circuit Technology, vols **1-4**, 2353-2356 (2008).
3. N Cherepy, B Sturm, O Drury; T Hurst. S Sheets, L Ahle, C Saw, M Pearson, S Payne, A Burger, L Boatner, J Ramey, E van Loef, J Glodo, R Hawrami, W Higgins, K Shah, W Moses, "SrI_2 scintillator for gamma ray spectroscopy ," Proc. SPIE, 7449, 7449-0 (2009); N Cherepy, S Payne, S Asztalos, G Hull, J Kuntz, T Niedermayr, S Pimputkar, J Roberts, R Sanner, T Tillotson, E van Loef, C Wilson, K.S. Shah, U Roy, R Hawrami, A Burger, L Boatner, W-S Choong, W Moses, "Scintillators with potential to supersede lanthanum bromide", Submitted to IEEE Transactions on Nuclear Science, IEEE Trans. Nucl. Sci., vol. 56, 873-880 (2009).
4. A.M. Conway, B. W. Sturm, L.F. Voss, P. R. Beck, R.T. Graff, R. J. Nikolic, A.J. Nelson and S.A. Payne, "Amorphous Semiconductor Blocking Contacts on CdZnTe Gamma Detectors," International Semiconductor Device Research Symposium, Dec 2009.
5. Y Dmitriev, L Cirignano, K Shah, "The effect of doping on X-ray response of TlBr crystals", Nuclear Instruments and Methods in Physics Research A **594** 206–209 (2008).